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Skull melting and top seeded solution growth of YVO ₄ were studied. A computerized optical spectroscopy system was designed and implemented. Laser pumped laser gain measurements were made of Nd:YAG and Nd:CAMGAR.		

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CRYSTAL GROWTH BY SKULL MELTING

by

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Final Report

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Final Report

Crystal Growth by Skull Melting

I. Crystal Growth

Skull melting of YVO_4 was thoroughly investigated except for experiments with large ($>5\text{kg}$) melts. This was not practical with our 20 KW rf generator. However, a double walled water cooled fused quartz skull holder was developed which was less lossy and far less expensive than the copper finger type sold by A. D. Little, Inc. No evidence of macrocrystalline formation was found, in any of the melts, and all showed evidence of oxygen dissociation and the presence of a second phase. Since growth of sizeable pieces of this soft, easily cleavable material in a skull melt seemed unlikely, efforts were made to develop a Czochralski technique, pulling from the skull. This proved impractical due both to the usual crust formation on top of the melt and to the basic chemical instability of the melt.

The latter problem was approached in two ways, first by fluxing the melt to obtain lower temperatures in a SiC. resistance heated furnace and then by constructing a sealed top seeded solution growth furnace permitting operation with up to three atmospheres of oxygen at reduced temperatures. Stable melts were not found, even at 1200°C and 3 atmospheres (30 psig) of oxygen. At lower temperatures the solubility of YVO_4 was too low to give reasonable yields and growth rates. Some small crystals were grown, but bubble formation at the crystal-melt interface prevented the growth of large, scatter-free material. A paper on this work is in preparation.

II. Computer-Controlled Spectroscopy System

Manual spectroscopy to determine emission cross sections, branching ratios and energy levels in solid state laser materials has often been difficult and not completely effective due to certain measurement problems. When an emission spectrum is measured, the spectral response of the measurement system must be taken into account. Branching ratios can be obtained by either numerical integration of the emission lines or by the use of formulae which assume a particular line-shape, usually a Lorentz profile. Unless the latter method is based on a complete accounting of the broadening mechanisms, it is no more than a convenient, but physically meaningless, method of classifying data. This could lead to erroneous results. It is necessary to perform computations to test and verify lineshapes and, if necessary, perform numerical integration. This becomes even more important when there are two or more closely overlapping emission lines. Thus the need for fast computational power is apparent.

As mentioned in a previous report, in the fall of 1977 a Data General Nova 3 computer was acquired at no cost to this contract. A stepping motor has been installed in a McPherson Model 213 scanning monochromator, and digital outputs of the computer have been interfaced to the motor drive circuitry. The Nova is equipped with A/D input channels, so that the detected fluorescent signal can be monitored. There are also two D/A output channels which are used for displaying stored data on an x-y plotter. Interactive software has been designed and successfully implemented which scans user-specified spectral ranges and stores data on diskette, automatically performing real time corrections

to account for the spectral response of the diffraction grating and photomultiplier being used. A program to generate x-y plots is operational. Computational algorithms such as curve-fitting, smoothing and integration have also been implemented. The system was tested by investigating the properties of Nd:YAG. While the question of the correlation of the radiative lifetime, stimulated emission coefficient and quantum efficiency was still not completely resolved, interesting new results were obtained. A complete report is available on this work (Crystal Physics and Optical Electronics Report #22, May 1979). It is also available as a Master's thesis. Work in this area is continuing under other sponsorship.

III. Laser Pumped Laser Gain Measurements: Comparison of Nd:YAG and Nd:CAMGAR

Solid-state laser materials can be evaluated using spectroscopic techniques, but even if such an evaluation indicates that the material can be lased, the definitive proof is actual lasing of the material. The stimulated emission cross section is the parameter which is difficult to measure with spectroscopic techniques; different values have been obtained with different techniques. For example, the values obtained by different authors for the stimulated emission cross section of the 1.06 μm transition in Nd:YAG differ by 50%.

The effective gain for the laser transition is determined by the stimulated emission cross section of the laser transition and by the cross section of reabsorption of the laser radiation. The laser end pumping technique was studied as a method for lasing optically pumped solid-state laser materials in the cw mode and for measuring the effective cross section of the laser transition. This technique is applicable if the absorption coefficient at the pump wavelength is small; if not, transverse pumping has to be used. We found that for Nd:YAG and Nd:CAMGAR an argon ion laser at the 514.5 nm wavelength is a better laser pump than a cw dye laser.

For a given laser pump, losses and mirror reflectivities, the laser rod length can be optimized to obtain laser threshold operation with minimum absorbed pump power. For example, with the argon ion laser as a pump, high mirror reflectivities and zero diffraction losses, the optimal length for Nd:YAG and Nd:CAMGAR operated at the 1.06 μm transition is 0.83 cm. For Nd:CAMGAR operated at the 0.94 μm transition it is 0.26 cm.

To reduce the power absorbed required to obtain threshold operation, a cavity with small diffraction losses is desired. The heat dissipated by the pump beam produces thermal distortion that can lead to large diffraction losses of the plane-parallel, concentric and hemispherical cavities, because they are in the boundary that separates the stable from the unstable cavities. The large diffraction losses of the plane-parallel cavity are reduced by a positive thermal lens. The diffraction losses of the concentric and hemispherical cavities are increased by positive thermal lensing. For the confocal cavity thermal lensing is not as important because this cavity is in the middle of the stability region. It was found that the confocal cavity had negligible diffraction losses.

An additional variable that can be changed is the laser beam radius in the laser rod. The population inversion is proportional to the pump power absorbed per unit area. Therefore, if the cross section of the laser beam is decreased, a larger population inversion can be obtained.

The laser end pumping technique requires overlap of pump and laser beams in the laser rod. The method used to obtain the closest overlap of the two beams is by tilting the cavity mirrors in order to maximize the laser power output for a given pump power input. Tilt of the cavity mirror produces a change in orientation and position of the laser beam. Therefore, the fraction of the laser beam volume on which there was overlap with the pump beam in the laser rod will change. The laser power output will increase if the overlap is better and decrease if worse. On the other hand, the mirror tilt also changes the alignment of the laser cavity itself. Mirror misalignment causes an increase in the diffraction losses and, therefore,

a reduction of output power. We need a cavity for which a close overlap of the two beams can be obtained and with tolerance for mirror misalignment.

The cavity that best satisfies these two requirements is the confocal cavity. It is the cavity with the highest tolerance for mirror misalignment. By tilting the cavity mirrors and rotation of the cavity it is possible to align the beams. It was not possible to rotate the confocal cavity with the experimental set-up used, so that for this case the plane-parallel cavity was better than the confocal because the position of the laser beam is determined by the pump beam. For future work a rotatable confocal cavity should be used.

Nd:YAG was lased at $1.06 \mu\text{m}$ and Nd:CAMGAR at $1.06 \mu\text{m}$ and $0.94 \mu\text{m}$, using as a pump an argon ion laser operated in the cw mode at the 514.5 nm wavelength. Both materials were lased in the cw mode at room temperature. The cross section of the $1.06 \mu\text{m}$ transition was measured from the slope of the line fit of the pump power absorbed at threshold versus $-\ln R_1 R_2$, where R_1, R_2 are the effective mirror reflectivities. For Nd:YAG the cross section was $\sim 2.0 \times 10^{-19} \text{ cm}^2$ and for Nd:CAMGAR it was $\sim 3.0 \times 10^{-20} \text{ cm}^2$ at $1.06 \mu\text{m}$.

These cross sections are smaller than those obtained from spectroscopic measurements. The following processes might have been the cause for this behavior:

1. Reabsorption of the laser radiation.
2. Absorption of the pump radiation by the $^4F_{3/2}$ manifold.
3. The cross sections were obtained assuming that 100% of the pump power absorbed was used to build up a

population inversion in the laser beam volume. Since the two beams are not perfectly aligned, only a fraction of the pump power absorbed will be actually used to build up the population inversion in the laser beam volume. As a consequence of this loss, the cross section will have a smaller value than with perfect alignment. Misalignment of the two beams is the largest source of uncertainty in the measurement of the cross section.

Nd:CAMGAR was lased at $0.94 \mu\text{m}$ with a plane-parallel cavity (only plane mirrors were available). Using almost the maximum power output of the argon ion laser, the material was lased with high reflectivity mirrors. The value of the cross section obtained from this experiment was $\sim 6 \times 10^{-20} \text{ cm}^2$.

In Nd:YAG and Nd:CAMGAR, the upper laser level of the $1.06 \mu\text{m}$ and $0.94 \mu\text{m}$ transitions is a level in the $^4F_{3/2}$ manifold. Therefore, competition between the two laser transitions will occur. The cross section of the $0.94 \mu\text{m}$ transition in Nd:YAG is $1.9 \times 10^{-19} \text{ cm}^2$, smaller than the cross section of the $1.06 \mu\text{m}$ transition. Therefore, a mechanism has to be provided to selectively increase the losses at $1.06 \mu\text{m}$ if lasing at $0.94 \mu\text{m}$ is desired. On the other hand, for Nd:CAMGAR we found that the cross section at $0.94 \mu\text{m}$ is bigger than at $1.06 \mu\text{m}$. Since this measurement was only made on the basis of one experiment, the errors were quite large. Spectroscopic measurements give a value of $3.2 \times 10^{-20} \text{ cm}^2$ for the $0.94 \mu\text{m}$ transition, which is approximately equal to the cross section at $1.06 \mu\text{m}$. A definite

conclusion cannot be reached, but if the cross section at $0.94\mu\text{m}$ is indeed larger than at $1.06\mu\text{m}$, then it would not be necessary to have a mechanism to selectively increase the losses at $1.06\mu\text{m}$.

A masters thesis has been written on this work.